FRICITION AND WEAR OF CARBON-CARBON COMPOSITE
PART 1: FRICTION TRANSITION AND STRUCTURAL CHANGES OF FRICITION FILM

Kia-Moh Teo and Khalid Lafdi
NSF-University-Industry
Center For Advanced Friction Studies,
Southern Illinois University at Carbondale, IL. 62901-4343, lafdi@siu.edu

Literature review
Carbon-carbon composites are commonly used as friction materials because of their exceptional thermal and mechanical properties. Unlike most of frictional and structural materials, C-C composites are extremely stable at high temperature.

Stimson et al. discussed in detail the design fabrication and engineering of carbon brakes [1]. However, until today the manufacturing of carbon-carbon composite brakes remains very costly and time consuming.

When two carbon materials are rubbed together, friction and wear properties are always very predictable. For given conditions, they might have a very significant change. Friction transitions have been observed in many which the value of the coefficient of friction changes drastically. Many published articles show the variation of the coefficient of friction in regard to time, pressure and temperature. Blau subdivided such transitions into eight stages [2]. But, the variation of friction coefficient is often subdivided into three regions: run-in, mild wear and transition to severe wear. Many models were developed to predict the variation of the coefficient of friction as a function of time. Some of these models could predict the run-in time of a bearing surface and checking the reproducibility of a surface contact condition [3]. Some models have great success in predicting the average of coefficient of friction. However, the term average friction coefficient is very misleading because of the presence of friction transitions. Asperity models based on mechanical deformation have also been developed but many friction details are neglected such as the effect of debris, material compositions and other surface reactions. Kuhlmann-Wilsdorf’s model that relates the coefficient of friction to asperity properties, shear stresses and microstructures is not sufficient to cover the actual sliding conditions [4]. Transition phenomena at the surface have been described as combination of multiple effects such as subsurface fatigue damage, wear-through of surface film, build-up of transfer layers, agglomeration of debris and change of material properties by Blau [1]. He also mentioned that the magnitude of the friction coefficient during transition is proportional to the formation of wear debris.

Many experiments were carried out to study the effects of load and speed on the wear and friction profiles. Friction transitions were found dependent on sliding energy [5]. In some hybrid semi-metallic materials, friction transitions were attributed to the formation and the break down of the oxide layer on the friction surface [6]. Studies on the structure of friction film, its formation could be one of the ways to understand friction and wear mechanisms [5]. Other factors such as contaminating or humid environment (humidity and oxygen) are believed to affect severely the friction and wear process. In addition, various contaminants may also alter wear and friction processes [7]. It was first encountered in the late 1930s that in the case of aircraft commutators made of carbons, wear always increased at high altitude because of the gas environment and lack of water vapor in the atmosphere [7]. Water vapor appears to have a lubricating effect on non-graphitic carbons [8]. It was found later that the coefficient of friction of graphite remained low only in the presence of water or other condensable vapors [9-11]. Indeed, the friction coefficient of carbon was shown ten times higher in vacuum. Therefore, the low coefficient of friction of graphite is not an inherent property. Bryant et al. showed that in vacuum the inter-laminar bonding of graphite is high [12]. This inter-laminar bonding decreased in the presence of air or water. They also suggested that water might reduce the binding energy of graphite. Rowe observed a similar effect but offered an alternative mechanism [13]. He suggested that water or oxygen might intercalate between the carbon basal planes and reduce the pi electron bonding. However, the work of Arnell and Teer didn’t show any increase in d interlayer spacing [14]. Their work contradicts the assumption of water molecule intercalation between the graphene layers. Yen and Ishihara published some surface studies on friction of carbons [15]. They showed that during severe friction conditions, two types of surface morphology are present: a dull-looking gray surface and mirror-like black surface, which they ascribed to contact pressure alterations. The contact pressure is higher at the gray area, which leads to a rougher surface without continuous wear debris film coverage. They also concluded that these contact pressure differences are caused by non-uniform thermal expansion due to frictional heating. Li and Sheehan found that friction and wear decrease with increasing temperature to 950°C due to chemisorption of impurities to form lubricating film, and to oxidation [16].
Experimental procedures

A sintered carbon composite was prepared for the friction test. The tests were performed using a standard test machine (Friction Assessment Screening Test “FAST”) purchased from LINK Engineering Company. All samples were tested for 90 minutes under various constant frictional forces in which only the rotational velocity was varied. A K-type thermocouple embedded in the middle of the sample at 2.5 mm from the sliding surface recorded the temperature history of the entire test period. In situ video capturing of surface evolutions and visual examinations were also conducted to understand the development of the friction film and its relationship to friction transition.

All carbon samples before and after the tests were characterized at different scales using optical microscopy (OM), surface profilometry, scanning electron microscopy coupled with Edax analysis (SEM EDAX), transmission electron microscopy (TEM), atomic force microscopy (AFM) and scanning tunneling microscopy (STM). Wear was computed from the thickness and the mass of the sample, which were measured before and after a test.

Fabrication of the carbon composite

The carbon composite has been made of compacted anisotropic coke and bonded with phenolic resins. The samples are fabricated in the cylindrical mold in which a pressure is applied. After hot pressing, the materials were carbonized, in flowing N₂ gas at 1000 °C. Graphitization was performed by raising the furnace temperature 4°C per minute until 2200 °C was reached. The furnace was held at this temperature for thirty minutes.

Friction assessment screening test “FAST”

The Friction Assessment Screening Test (FAST) machine (Figure 1) was developed by Ford Motor Company specifically for rapid “fingerprinting” of friction material specimens. The essential elements include a sample arm, which clamps against a rotating cast iron disc. This disc is attached through an insulated hub to the shaft of a 900rpm, 1 hp induction motor. The disc is 7 inches in diameter and 1.5 inches thick, and is designed so that either surface may be used. The design of the attachment also allows for the quick changing of discs. However, in this study the machine setup was modified to accommodate a 6-in. carbon rotor disk.

The lining sample mounts in a recess near the free end of the pivoted sample arm. The clamping load is applied by hydraulic pressure acting on a rolling diaphragm load cell. The connection between the clamping assembly and the sample arm is through a flexible tension member, and the load system is designed to reduce hysteresis and run-out. The load control system incorporates a control valve assembly, which allows tests to be performed either at constant friction force (output), or constant friction clamping load (input). The friction force is transmitted directly to the friction valve spool. In the constant output mode, any variations of the friction force results in a displacement of the clamping valve spool. The pressure automatically adjusts to re-establish the original equilibrium. The magnitude of the friction force may be selected by means of the load control screw.

In the constant friction force mode, the clamping pressure is controlled to change at a rate inversely proportional to the change of friction coefficient. Thus, the recorder produces a plot of the reciprocal of the friction coefficient versus time.

To avoid recording high-frequency friction fluctuations during a test, the recorder has a frequency response limited to 10 Hz. Quality control (QC) tests generally run for 90 minutes.

Table 1 shows the testing conditions performed. Tests 1 to 6 of various rotational speeds were used to generate various total energies absorbed per minute. The frictional force was kept constant and adjusted to a value of 69.7 N. From test 7 to 12, various rotational speeds and a frictional force of 104.6 N were used to study the effect of higher frictional forces.

Results and discussions

Figure 2a shows the experimental results for the friction force of 69.7 N with various rotational speeds. All curves clearly show the sudden changes in the friction coefficient. Figure 2b shows the friction coefficient curves filtered with Gaussian Kernel smoothing equation as shown below.

\[ K \mu_i = \frac{\sum_{j=1}^{n} M \left( \frac{t_j - t_i}{b} \right) \mu_j}{\sum_{j=1}^{n} M \left( \frac{t_j - t_i}{b} \right) } \]

where

\[ M(s) = \frac{1}{\sqrt{2 \pi} \cdot 0.37} \cdot \exp \left( \frac{-s^2}{2 \cdot 0.37^2} \right) \]

and b is a bandwidth parameter for smoothing. The bandwidth used in the smoothing of the FAST result was 100. Using this equation, the vibration in the coefficient was successfully filtered with the trend maintained. However in some cases, the value of the coefficient of friction overshot the maximum value in the experimental result.

Depending on the testing conditions, the coefficient of friction profile as a function of time exhibits either single or multiple friction transitions (increase of coefficient of friction from the base line). Figure 3a shows a typical trend in friction coefficient during friction transition. As shown in Figure 3b, the coefficient of friction initially has a value of ~0.3 and as the test progressed with time the coefficient of friction decreases to a value of 0.16. The coefficient of friction is stabilized for a short period at point of 0.16. Then, the transition occurs and the coefficient of friction increases up to a value of 0.6. When the transition is over, the coefficient of friction begins to decrease. The same pattern may be repeated many times. At the beginning of the test, the bulk temperature (2.5 mm
from the surface) increases rapidly from the room temperature to a value of 200 °C within few seconds during run in process (it depends on testing conditions, the rate of temperature increase can exceed 10 °C/sec). Then, the temperature increases gradually at a rate of 0.05 °C/sec. After that, it remains steady at about 290 °C. However, at the first glance of the transition, a fast increase of bulk temperature is recorded with a rate between 0.2 – 0.45 °C/sec. After the transition, the temperature continues to increase very slowly due to the gradual wear of the sample.

Characterization of friction surface at different stages
Friction surfaces for all experiments exhibit some specific characteristics in which they can be summarized as follows:

a. Friction surface before friction testing
The samples are made of sintered needle coke flakes, carbonized and graphitized to heat treatment temperature of 2200 °C. In this case the samples consist of graphitic particulates and voids.
Before each friction test, both the sample and the rotor disk surfaces are sanded using sandpaper up to the grid size of 800. For all experiments, same surface finish is managed.
Also, the surface profile is taken before and after each test. As shown in Figure 4 & 5, the surface is relatively smooth with some visible voids.

b. Friction surface before friction transition
As the friction test progressed, friction film was built up (Figure 6a and b). The details of the coke grains were masked. Visual examination shows that the entire friction surface appears very shiny. In addition the surface topography appears very smooth with very tiny waviness (Figure 6c).

c. Friction surface at the friction transition
At the first indication of the transition, the coefficient of friction starts to increase and some surface alterations are observed as shown in Figure 7a. As shown at low magnification (Figure 7b), the friction surface of the sample appears to be made of two distinct bands or wear tracks: one is shiny (i.e. smooth) with some relative waviness and the second is dull with some random surface irregularity (i.e. rough). For comparison, surface topographies at the smooth and rough areas were performed (Figure 8). During the friction transition, dull slits (powdered wear track) appear with localized roughness, cracks and voids as shown in Figure 9.
On the other hand, the smooth part was observed to have polished bands and/or bands with some granular structure. This smooth part was made of a smeared friction film in which agglomeration of tiny particulates has occurred as shown in Figure 10a. These particulates with narrow range of sizes and shapes are highly compacted. As shown in both images (Figure 10b and c) the structure of the smeared film is made of low order carbon. The selected diffraction pattern showed a very diffuse halo (inset of Figure 10b) and we observe that few graphene layers remain unchanged (arrows in Figure 10c). Indeed, there is a structural change from very straight and anisotropic structure in form of graphitic structure into quasi-less crystalline and amorphous type structure. We believe that the degree of compaction is strongly dependent on the testing conditions (i.e. normal load, time and initial velocity). However, this smeared friction film might often be disrupted and contain cracks.

Characterization of the friction film using the Atomic Force Microscopy (AFM) in both topography and lateral force microscopy mode (LFM mode) shows that the friction film is very shiny along the wear track and it is relatively smooth (Figure 11a). The average roughness is about 42 Å. Since the surface roughness is smaller than 200 nm, the lateral force microscopy mode can be used to see if there is any friction coefficient gradient within this smooth friction film. We used Lateral Force Microscopy (LFM) to measure the frictional forces resulting from the AFM probe scanning over a sample. When the probe crosses an area of the sample, which has a higher drag or friction force, the cantilever will display a greater amount of torque. The opposite is true for areas where lower frictional forces are generated. Here 'high' and 'low' are relative terms, for instance an adhesive or charged particle on a carbon surface may exhibit a 'higher' frictional force than the surrounding substrate. Thus, the particulates appear brighter in the phase image. Below that, where the particulate could be said to be 'slippery' (having a low frictional coefficient), the particulate appears darker in the friction image. As shown in the LFM image (Figure 11b), we observed that the surface is made of narrow range of gray scale of brightness. The bright area means that the area has a higher friction property than the dark area.
Scanning of an area of 2 × 2 µm shows that the size of the particulates are in the range of 100 nm. Their shape appears to be elliptical, elongated or spherical particulates (Figure 11b). However the LFM images show that most of these particulates were slightly covered by a bright thin layer which indicates that this layer located on the skin of these particulates is stiffer and presents higher coefficient of friction than the dark part located on the core (Figure 11b). These AFM data confirmed the TEM observations from our previous works. It seems that a rolling mechanism has a great effect on the friction of carbons.

d. Friction surface as the friction transition progresses and after
During the transition, the powdered film expanded to reach the entire rubbing surface. The sheared particulates tend to form very compacted and smeared particulates with a narrow range of sizes and shapes in which the bonding force appears to be very large. At some locations, the interfaces between
particulates can be observed without any apparent gap between them even at nanometric scale. However, the dull part of the friction surface is clearly made of disrupted film particulates. For given testing conditions, the dull areas tend to expand and the surface becomes rougher in comparison to the shiny part. The number and the width of these dull tracks are strongly dependent on the testing conditions (normal load, time and velocity) and gas environment (argon, humidity and air). At some locations in the dull area, cracks and holes are very obvious. The cracks are either empty or filled with wear debris. The extension and the magnitude of these bands and their shapes depend on many parameters (oxidation, thermal expansion, thermal stresses and others). However there is always a fair matching at macro-metric scale between friction surfaces of the rotor and stator (Figure 12).

After the transition, the sliding surfaces gained back the same surface characteristics and properties. It consists mainly of very shiny surface with fine particulates.

Conclusions

In general, we found that the evolution of friction transition is closely related to the energy absorbed and friction film. During transition, friction film was disrupted following the creation of wear tracks that were made up of debris. Friction surfaces of the sample were examined using various techniques. We observed that a structural change from graphitic to amorphous carbon had occurred. Wear tracks created at the transition stage were very porous due to surface damage by debris and oxidation.

The classical explanation for the low friction of graphite relates friction to the weak binding energy of graphite lattice, therefore little energy is needed to induce the cleavage or shear, and so the coefficient of friction is low. However this easy cleavage is true only at wet and condensable vapors are present in the environment. However if there is any substantial rise in surface temperatures, the water saturation decreases and the carbon surface is physically desorbed, leading to an increase of coefficient of friction. High coefficient of friction and wear may also be induced at room temperatures if the combination of load and velocity produce an increase of surface temperature in the order of 600 °C or greater. A part from the water effect, they are other factors that influences the friction transition and the magnitude of the coefficient of friction. Finely divided debris from the wear process that forms the friction film is made of nanometric amorphous sheared particles. This friction film may enhance the preferred crystallographic orientation and direction of easy shear of carbon crystallites or rolled particulates. Midley and Teer have observed that the latter effect occurring in a cyclic manner during the sliding of carbon throwing out (1963). When the friction rises to higher value, the surface stresses become sufficient to disrupt the layer-consolidated debris. Then, the surface is roughened and the real contact area decreases. This leads to a sudden increase in friction coefficient. However, we observed an increase of wear debris that leads to an increase of coefficient of friction through inter-laminar shear of particulates formed from the wear debris. This hypothesis and the extended correlation of testing parameters, wear and surface properties will be illustrated further.

References

8. Ramadanoff and, D. and Glass, S.W., 1944, Trans. AIEE, 6, pp. 825.
Figure 1. The Friction Assessment Screening Test (FAST) machine.

Table 1. Testing conditions

<table>
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Figure 2. Friction results for various power input conditions (a) Friction coefficient versus time (b) Filtered friction coefficient versus time using the Gaussian Kernel smoothing technique.
Figure 3. (a) Model illustrating multiple stages of the coefficient of friction variation as a function of time (regardless the velocity and power input values); (b) Variation of coefficient of friction and temperature as function of time for a velocity of 4.24 m/sec and frictional force of 69.7 N.

Figure 4. Surface of the sample before tests (a) Image of the entire sample before friction test; (b) Magnification of the circled area; (c) Magnification of the squared area;

Figure 5. Surface profilometry of the sample before the test
Figure 6. Surface of the sample before the friction transition (a) Image of the surface of a sample before the friction test; (b) Image of the friction surface before the friction transition; (c) Surface topography of the sample after the friction test and before the friction transition

Figure 7. Surface alteration at the start of the friction transition (a) Image of the friction surface at the first glance of friction transition; (b) Low magnification of the powdered slit (indicated by the arrow)

Figure 8. Surface topography of the friction surface at the transition stage. Smooth part of the surface at transition stage (double arrows in Figure 8); Rough part of the surface at transition stage (single arrows in Figure 8)
Figure 9. some SEM images of the powdered wear track (dull slit) at the beginning of friction transition.

Figure 10. (a): High magnification SEM images of the smeared friction film, (b) Bright field TEM image of the smeared friction film with the diffraction pattern of the smeared film at the lower left corner; (c) High resolution TEM image (Lattice fringe of TEM modes) of a single particle of the corresponding smeared film.
Figure 11. AFM characterization of the friction film (a) Topography image (b) LFM image of the friction film.

Figure 12. Surface topography matching of the rotor and the stator.